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D E C I S I O N
of 17 July 2001

Case Number: T 1061/98 - 3.3.3

Application Number: 94113247.4

Publication Number: 0640641

IPC: C08G 69/10

Language of the proceedings: EN

Title of invention:
Process for preparing polysuccinimide

Applicant:
Mitsui Chemicals, Inc.

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Obviousness (no) - special reaction mechanism"

Decisions cited:
-

Catchword:
-

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Case Number: T 1061/98 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 17 July 2001

Appellant: Mitsubishi Denki Kabushiki Kaisha
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Decision under appeal: Decision of the Examining Division of the European Patent Office posted 9 June 1998 refusing European patent application No. 94 113 247.4 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. Young
Members: P. Kitzmantel
J. C. M. De Preter

Summary of Facts and Submissions

I. This appeal, which was filed on 14 August 1998, lies against the decision of the Examining Division dated 9 June 1998, refusing European patent application No. 94 113 247.4 filed on 24 August 1994 in the name of MITSUI TOATSU CHEMICALS, INC., later merged into MITSUI CHEMICALS, INC., claiming a JP priority of 24 August 1993 and published under No. 0 640 641. The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 13 October 1998.

II. The decision under appeal was based on sets of each eight claims of a main and an auxiliary request, both filed with a submission dated 27 April 1998, Claim 1 of the main request reading as follows:

"1. A process for preparing polysuccinimide by dehydrating condensation of aspartic acid in an organic solvent comprising removing a portion or more of the organic solvent from the reaction mixture while charging to the reaction mixture a new solvent or an additional organic solvent which is treated by a drying agent, distilled or dried by using other solvents to reduce the water content, wherein the new or additional solvent has a water content of 50 ppm or less, which is smaller than that of the removed organic solvent."

Claims 2 to 8 of the main request were dependent on Claim 1.

The auxiliary request differed from the main request only by deletion from Claim 1 of the feature "or dried by using other solvents".

III. The decision under appeal held that the subject-matter of Claims 1 of both requests was obvious over the teaching of document

D1: FR-A-2 403 353,

which disclosed the preparation of polysuccinimide by polycondensation of aspartic acid under the conditions of azeotropic dehydration, because the skilled person was aware that by further reduction of the water content of the reaction mixture polymers of higher molecular weight could be produced.

IV. With its submission dated 18 June 2001, filed in response to the Rapporteur's communication of 8 March 2001, the Appellant submitted as its sole request an amended set of seven claims, Claim 1 reading as follows:

"1. A process for preparing polysuccinimide by dehydrating condensation of aspartic acid in an organic solvent comprising removing a portion of the organic solvent and water generated in the reaction process from the reaction mixture together with the organic solvent used while charging to the reaction mixture

(i) the organic solvent which was distilled off with water after having been treated with a drying agent or distilled to reduce the water content or

(ii) a new organic solvent containing a lower amount of water than the organic solvent distilled off,
wherein the charged organic solvent has a water content 50 ppm or less."

V. In its written submissions and at the oral proceedings held on 17 July 2001 the Appellant argued that the claimed subject-matter was non-obvious over D1, because the skilled person would not have expected that polysuccinimide of higher molecular weight could be obtained by substituting solvent having a water content of 50 ppm or less for the solvent which is drawn off together with the reaction water by azeotropic dehydration.

The "law of mass action" would not support the conclusion that a further reduction of the water content in the reaction mixture would necessarily shift the equilibrium to polymers of higher molecular weight, because

(i) polyimides were more resistant to hydrolysis than polyesters,

(ii) the polymer forming step of the preparation of polysuccinimide from aspartic acid, i.e. the formation of polyamic acid, did not involve the elimination of water, and because

(iii) even if the imide formation reaction was promoted by the reduction of the water content, this could by same token either lead to many short polymer chains or to fewer long polymer chains.

With regard to afore-mentioned point (ii) the Appellant submitted at the oral proceedings a scheme of this reaction (cf. point 4.4.2 below) and, in support thereof, a copy of the Article "Chemical Studies of Polyaspartic Acids" by J. Kovacs et. al. (hereinafter document D2) as set out on pages 1084 to 1091 of volume

26 (even pages) of an unidentified source, the odd pages of which indicate the date "April 1961", showing in the Appellant's submission that said Article had been published before the priority date of the application in suit.

As an additional argument the Appellant pointed out that the prior art process for the preparation of high molecular weight polysuccinimide, which was known from US-A-5 142 062, a document filed about two years before the present priority date and acknowledged in the introduction of the application in suit, was very cumbersome in that it comprised a first polymerization step in the presence of a phosphoric acid catalyst to a solid intermediate, which had to be comminuted, further polymerized to the final degree of polymerization and finally purified. In the Appellant's view, this document illustrated the non-obviousness of the present simple method for the preparation of high molecular weight polysuccinimide.

VI. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of claims 1 to 7 filed with the submission dated 18 June 2001.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments (Article 123(2) EPC)*

Claim 1 is based on its original version, on the statements on page 5, lines 14 to 18 (features (i) and (ii)) and on original Claim 6 (maximum water content of charged organic solvent).

Claims 2 and 4 to 7 are, respectively, based on original Claims 3 and 7 to 10; Claim 3 is supported by the statement on page 6, lines 17 to 19 of the original application.

The requirements of Article 123(2) EPC are therefore complied with by all claims.

3. *Novelty (Article 54 EPC)*

3.1 Document D1, whose US counterpart US-A-4 363 797 is acknowledged on page 2, lines 14 to 17 of the application in suit, discloses several methods for the preparation of polysuccinimide (= polydehydroaspartic acid: formula II) (page 3, lines 18 to 29; page 4, line 24 to page 7, line 31). According to the preferred method (method D) of D1 aspartic acid and a strongly acidic ion exchange resin are heated in a solution of diphenylether and the water formed during this polycondensation reaction is azeotropically drawn off via a Dean-Stark distillation trap (page 5, lines 8 to 16; page 7, lines 1 to 31).

3.2 In view of the information by the Applicant (page 2, 4th paragraph of submission dated 2 December 1997; point 3 of Reasons of decision under appeal) it is accepted by the Board that according to D1 the water content of the solvent (diphenylether) cannot drop below about 100 ppm.

3.3 The subject-matter of present Claim 1 is, thus, novel over the disclosure of D1.

4. *Problem and solution*

4.1 The problem underlying the claimed subject-matter with respect to D1 is the development of a process for polycondensing aspartic acid to polysuccinimide having a molecular weight in excess of 10.000 with simple procedures (page 3, lines 4 to 14 and 21 to 23 of the application in suit).

4.2 According to Claim 1 this problem is solved by substituting organic solvent having a water content of 50 ppm or less for the solvent which is drawn off in the course of the azeotropic dehydration of the polycondensation reaction.

4.3 The available evidence, which covers the claimed embodiments (i) (Examples 1 to 4 of application in suit; Example 1 and Comparative Example 1 of the submission dated 27 April 1998) and (ii) (Example 2 and Comparative Example 2 of the submission dated 27 April 1998), confirms that by the claimed method molecular weights may be achieved, which are considerably above 10.000.

According to this experimental evidence the polymeric product is recovered from the reaction mixture by filtering (Examples 1 to 3 of the application in suit; Example 1 of the submission dated 27 April 1998) or precipitiation with a non-solvent (Example 4 of the application in suit; Example 2 of the submission dated 27 April 1998), washing and drying.

It is, thus, accepted that the existing technical problem has effectively been solved by the subject-matter of present Claim 1.

4.4 Obviousness

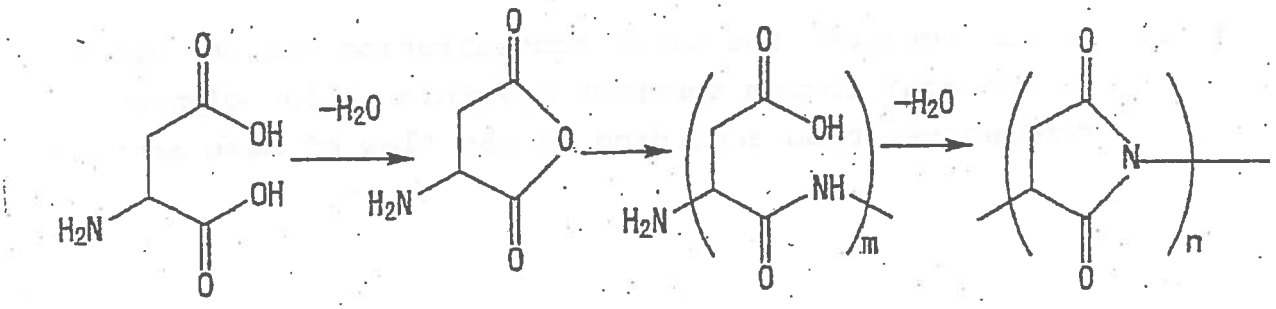
In view of the arguments brought forward by the Appellant (cf. point V supra) the Board is satisfied that the claimed subject-matter involves an inventive step.

This conclusion is based on the following considerations:

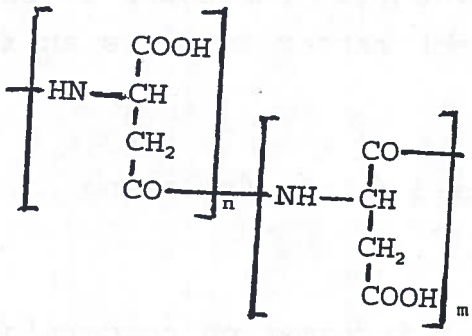
4.4.1 D1 contains no explicit information concerning the relationship between the molecular weight of the polysuccinimide and the water content of the solvent.

It only mentions in connection with methods C and D (page 6, line 25 to page 7, line 17) that the water which is formed during the polysuccinimide formation is distilled off via a Dean-Stark trap.

4.4.2 While the decision under appeal was right in stating that in the case of "ordinary" polycondensation reactions (e.g. the formation of polyamide-66 from adipic acid and hexamethylene diamine) a decrease in the amount of water in the reaction mixture will result in an increase of the molecular weight of the obtained polymer (cf. point 4.3 of the Reasons of the decision under appeal), the formation of polysuccinimide from aspartic acid is not such an "ordinary" polycondensation reaction, but, according to the Appellant (cf. point V supra), involves the following more complicated reaction sequence:



The above reaction scheme and particularly the intermediate formation of a polyamic acid (poly-α,β-L-aspartic acid) of the formula (III)



(III)

is confirmed by the disclosure on page 1085, left hand column of D2.

In view of the printed format of D2 and of the header information it carries ("vol. 26"; "April 1961") the Board sees no reason to doubt the public status of the relevant information it contains. Moreover, it appears that D2 corresponds to the document "KOVACS et Coll; J. Organic. Chemical. 26, 1081 (1961)" referred to on page 4, lines 29 to 32 of D1, not only because of the congruence of the bibliographical data, but also because the reference in D1 fits the experimental data in the right hand column of page 1087 of D2.

4.4.3 It is evident from this reaction scheme that the formation of the polymer chain which eventually defines the molecular weight of the final polymer is not linked to the water concentration of the reaction mixture, because this step does not involve any dehydration.

4.4.4 In consequence, the water concentration has no impact on the equilibrium constant governing this polymer forming reaction according to the "law of mass action"

and the skilled person had no reason, therefore, to expect any influence of the amount of water in the reaction mixture on the molecular weight of the resulting polymer.

4.4.5 The objection of lack of inventive step over D1, on which the decision under appeal was based, cannot, therefore, be maintained.

4.4.6 In view of the afore-mentioned experimental data (cf. point 4.3 supra) the Board is also satisfied that the subject-matter of Claim 1 is non-obvious over the further known prior art, particularly over the disclosure of the US-A-5 142 062 (cf. point V, last paragraph supra), because the claimed method provides polysuccinimides of higher (as compared to D1) molecular weight by a method which is considerably simpler than the method according to said US-A which requires mechanical comminuting and further polymerization and finally purification of the solid reaction product.

4.4.7 There is no need, therefore, to assess the validity of the further arguments brought forward by the Appellant in support of its assertion that the "law of mass action" was not an appropriate basis for the Examining Division's allegation of a necessary interdependence of the amount of water in the reaction mixture and degree of polymerization (cf. points (i) and (iii) of paragraph two of Section V supra).

4.4.8 In the Board's judgment, therefore, the subject-matter of Claim 1 involves an inventive step.

4.4.9 By virtue of their appendancy to Claim 1 the same conclusion applies to Claims 2 to 7.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant the patent on the basis of Claims 1 to 7 filed with the submission dated 18 June 2001 after any consequential amendment of the description.

The Registrar: The Chairman:

E. Görgmaier R. J. Young